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Risk Assessment Exposure of Mercury (Hg) at People who Consuming Nila Fish (*Oreochromis niloticus*) from Limboto Lake of Gorontalo Province.

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ABSTRACT

This study objective was to risk assessment of mercury at people who consuming Nila fish (Oreochromis niloticus) from Limboto Lake of Gorontalo Province. Observational study was used by using environmental health risk assessment approach. Mercury levels in Nila fish (Oreochromis niloticus) from Limboto Lake analyzed with Atomic Absorption Spectrometry, whereas body weight, consumption rate and exposure time analyzed quantitatively with interviewing 95 participants from six village for calculated mercury intake and risk quotient (RQ). The mercury levels in Nila fish (Oreochromis niloticus) from Limboto Lake, approximately 0.000007 - 0.000089 mg/gram. This study found that the average of risk quotient at people who consuming fish in Payunga village with 0.0000554 mg/gram was 1.32, risk quotient for consuming fish in Podutuma village with 0.000061 mg/gram was 1.01, risk quotient for consuming fish in Ilomangga village with 0.000007 mg/gram was 0.11, risk quotient for consuming fish in Kayubulan village with 0.000067 mg/gram was 1.36, risk quotient for consuming fish in Huntulabohu village with 0.000089 was 2.87 and risk quotient for consuming fish in Buhu village with 0.000089 was 1.08. This study showed that fish not secure for consuming until 30 years later so that the required risk management. Risk management can be done by reducing levels of mercury in fish, controlling consumption rate, and lessening exposure time. However, this study suggests that the most effective risk management for managing risk is controlling consuming rate for fish from Limboto Lake. Keywords - Risk assessment, Mercury, Nila fish (Oreochromis niloticus), Limboto Lake

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INTRODUCTION

Gold mining in the province of Gorontalo long ago been traditionally managed by the community. Formally managed gold mining activities are not permitted by the government of society, both the provincial and district level. One of the most troubling issues for communities around the site of gold without a permit is the use of toxic materials (B3), namely: mercury (Hg). The use of mercury as an ingredient for binding and gold ore separator with sand, mud and water that is not properly managed will impact the gold miners and the community about the location of illegal mining, where mercury has been used from the management of gold ore usually dumped in water bodies and consequently the Limboto Lake outlet of the mercury waste disposal. Waters contaminated with mercury (Hg), may have an impact on aquatic biota that live and thrive in these waters, especially waters used for aquaculture. One of the waters used for aquaculture, the water of the lake. Lake water, including surface water, is the most polluted water sources as a result of human activities, fauna, flora and other substances (Chandra, 2007).

Heavy metals into the body of living beings in general are in several ways, namely, digestive tract *(ingestion),* breathing *(inhalation)* and contact with skin *(dermal).* Metal absorption through the gastrointestinal absorption of p through food is one of the important pathways of toxicant absorption. Many environmental toxicant go through the digestive tract. The absorption processes causes toxic effects unless it is absorbed by the body (Mukono, 2010). Through the respiratory tract *(inhalation)* Toxic effects are usually quite large, both in animals that enter the water through the gills, and land animals that go through the dust in the air to the respiratory tract. While metals that enter through the skin, toxic effects and absorption is relatively small. (Darmono, 2010).

Research conducted by Simbolon *et al,* in 2010, about the levels of mercury and cyanide in fish caught from the Gulf of Kao, North Halmahera. It is known that mercury levels are in two locations in the waters of the Gulf of Kao, relatively low at 0.001 ppm, while levels of mercury that accumulates in the body of the red snapper, which is in the heart, can reach 0.38 ppm and 0.19 ppm the meat section. As for the mullet, the mercury content in the liver to 0.36 ppm and 0.25 ppm reach the meat. Jackfruit seeding for fish mercury content in the liver reaches 0.61 ppm, and meat section, reaching 0.04 ppm. This suggests that the levels of mercury in fish red snapper, mullet, and at the heart of jackfruit seed fish, which was caught in the Gulf of Kao North Halmahera has exceeded the maximum limit of heavy metal contamination in food according to Indonesian National Standard in 2009 which is 0.5 ppm, (Simbolon, et.al, 2010).

Limboto Lake currently located on the condition that very concern because of the shrinkage and siltation due to sedimentation that threatens the future existence. In addition to the silting problem, various community activities around and in the lake region is also increasingly threatened and worsen the preservation of the lake. Currently Limboto lake water quality deteriorated due to domestic waste, Nila fish (Oreochromis niloticus) fish farming activities are carried out in the lake, lake sedimentation due to erosion in upstream areas.

The pollution caused by the accumulation of the inlet, sedimentation and agricultural activities around the lake. Heavy metals such as Cd, Hg, Pb, and Cr6^{+,} and Se, As, Zn, Fe, and Mn in the waters of Limboto Lake detected. Based on data from the Environment Agency, Research and Technology (Balihristi), Gorontalo Province in the year 2013, it is known that mercury levels in lake waters Limboto detected at that 5 point in the Batudaa village of Barakati subdistrict Hg concentration 0.0085 mg / L, in subdistrict Talaga Jaya Hg levels of 0.0082 mg / L, in the subdistrict of Talaga Biru Hg levels of 0.0089 mg / L, in the Hutadaa village of Hg levels 0.0288 mg / L and at Iluta village level 0,0117mg Hg / L. Of 5 points detected mercury Limboto lake waters, it is known that the water quality of Limboto Lake is above the maximum permissible levels for Class III water is 0.002 mg / L, based on Government Regulation No. 82 of 2001 on Water Quality Management and Water Pollution Control. (EART, 2013).

That it contains Mercury (Hg) in lake water Limboto, may indicate there has been a contamination on aquatic biota that live in it. Based on the catches of fishermen in Limboto lake In 2007 it is known that the highest fish catches in Limboto Lake, which catches fish of the genus Oreochromis niloticus Oreochromis species, where the catch of this kind as much as 154.1 tons, or by 24.3% (Resmikasari, 2008). This suggests that the fish catch is a fish species most consumed by the public.

Payunga Village, Podutuma Village, Ilomangga Village, Kayubulan Village, Huntulabohu Village, and



Buhu Village are the villages where located in the side of Limboto Lake. The people there are high fish consumption, which means that the people consume more fish than the other people who live in other villages around the Limboto Lake. This is because some people are fishermen and their houses are near with traditional market. Although some of them are fishermen, they also eat fishes that they buy in the market. Limboto Lake is very risky affected by heavy metal mercury (Hg) that may exist on the fish. Therefore, to anticipate the effects of mercury (Hg) in the future society, the study aims to know the health risks of heavy metal pollution of mercury (Hg) in the meat Oreochromis niloticus in Limboto Lake Gorontalo Province.

METHODS

This type of research using observational methods to design Environmental Health Risk Analysis in the form of observations in the sample to find a picture of the variables studied, namely exposure to mercury in Oreochromis niloticus. From the results of the fish samples were then analyzed using AAS (*Atomic Absorption Spectrophotometry*).

This study will be conducted in April-June 2014 around Limboto lake at 6 stations namely Payunga Village, Village Podutuma, Ilomangga Village, Village Kayubulan, Huntulabohu Village, and the Village buhu, Preparation tool in April 2014 Sample preparation Oreochromis niloticus weekly analysis conducted in the laboratory while the heavy metal concentrations of mercury (Hg) for fish and water samples and performed by AAS method (*Atomic Absorption Spectrophotometry*) in the Laboratory of Development and Fisheries Products Quality Testing (LPPMHP) Gorontalo Province.

The research environment in this population are all kinds of Oreochromis niloticus, lake water and Communities around Limboto lake who consume Nila fish (Oreochromis niloticus). The samples in this study are Oreochromis niloticus and Limboto lake water taken from the sixth station. Criteria for fish sampled in this study, namely Oreochromis niloticus weight <250 grams or tanggan palm-sized adults, while the lake water sample is at a depth of 2 meters.

The human population is a whole community around Limboto Lake who consume Oreochromis niloticus. Human samples in this study were community residing in 6 (six) research station as many as 95 people. Sampling was conducted using *proportional random sampling*.

This study uses Environmental Health Risk Analysis using the Excel program. Environmental health risk analysis carried out by the following procedure:

Calculating the intake using the formula:

$$I = \frac{CxRxf_ExD_t}{W_bxt_{avg}}$$

Specification:

/ (Intake): Total concentration risk agents that enter the human body with a certain body weight each day (mg / kg / day)
 C (concentration): Concentration risk agents in food (mg / kg)

R (Rate): The rate of consumption or intake of entering each day (gram / day)

 f_E (frequency of exposure): The length or the number of days of exposure per year (day / year)

 D_t (Duration time): The length or the number of years of exposure

W_b (Weight of body): Weight / population (kg)

 t_{avg} (average Time): The period of time the average day. 30 years x 365 days / year = 10,950 days (non-carcinogens). Or 70 years x 365 days / year = 25,550 days (carcinogenic).

Calculation of non-carcinogenic risk:

$$RQ = \frac{I}{RfD}$$

Specification

RQ : Intake which has been calculated on the formula 1 (mg / kg / day) RfD : Reference value at risk agent ingestion exposure

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RESULTS

Levels of Heavy Metals Hg (mg / kg) in 6 Statsiun Oreochromis niloticus in Limboto lake 2014

Village	Hg concentrations (mg / kg)	Standard (mg / kg)	Criteria
Payunga	0055	0.5	E
Podutuma	0061	0.5	E
Ilomangga	0007	0.5	E
Kayubulan	0067	0.5	E
Huntulabohu	0089	0.5	E
Buhu	0089	0.5	E

Table 1: Levels of Heavy Metals in Fish Hg at 6 Station in Limboto lake 2014

Source: Primary Data 2014

Note: E = Eligible, with quality standards according to SNI 2009

Table1 above shows that the levels of heavy metals Hg in Oreochromis niloticus in the higher concentration station in the Huntulabohu village and Buhu village of 0.089 mg/kg and the lowest levels llomangga village with levels of 0.007mg/kg, from 6 to station in Limboto Lake been contaminated with mercury, but still eligible or below the value of the maximum contaminant limit of mercury in fish by the Indonesian National Standard 2009 of 0.5 mg/kg. Hg content in Oreochromis niloticus could cause the content of Hg in lake water Limboto already exceeding quality standards and Oreochromis niloticus is included in the class oromnivores eating everything, so it occupies the high esttrophic waters (Setianto, 2011). Mercury in to the body Oreochrochromis niloticus, can be derived from water that has been contaminated with mercury, consumption sediment, aquatic plants, and small fish. Through the food line, metallic mercury enters through two ways, namely through the water (beverages) and plants (groceries) (Palar, 2008).

Levels of Heavy Metals Hg (mg / kg) in Water at 6 Statsiun in Limboto Lake 2014

Village	Hg concentration (mg / L)	Standard (mg / L)	Criteria
Payunga	0.0002	0.002	E
Podutuma	0.0020	0.002	E
llomangga	0.0023	0.002	NE
Kayubulan	0.0025	0.002	NE
Huntulabohu	0.0006	0.002	E
Buhu	0.0006	0.002	E

Table 2: Levels of Heavy Metals in Water at Station 6 in Limboto Lake 2014

Source: Primary Data 2014

Note: E= Eligible, with quality standards according to Indonesian Government Regulation no. 82 of 2014 NE =Not Eligible

Table 2 shows the heavy metal content of lake water Limboto to the highest levels of Hg in the village Kayubulan is 0.0025 mg/L and the lowest in the Buhu village and Huntulabohu village is 0.0006 mg/L. There are two villages that do not eligible according to Indonesian Government Regulation no. 82 of 2001 of the Ilomangga village and Kayubulan village. The presence of mercury in waters of the lake Limboto, can be derived from the natural elements contained mercury in nature either in stone or soil, the presence of household waste from settlements around the lake or river flow activity along an inlet of the lake, and the sediment carried by the river, Limboto into the lake, so that the waters of the higher levels of mercury and lead levels of mercury in waters of the lake Limboto had not qualified. This is supported also by the silting that occurs in Limboto Lake.

Intake

 Table 3: Intake (I) Consumption Oreochromis niloticus Containing Heavy Metals Hg with Exposure duration (Dt) 30 Year

 of the Community around Limboto Lake

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Metal Type	Location	Mean	Min	Max	SD
Intake/ I (mg / kg / day)					
Hg	Payunga	0.000140016	0.0000436	0.0003983	0.0001050478
	Podutuma	0.000100560	0.0000788	0.0001544	0.0000289536
	Ilomangga	0.000011711	0.0000074	0.0000244	0.0000054417
	Kayubulan	0.000137981	0.0000629	0.0002884	0.0000550597
	Huntulabohu	0.000368241	0.0000542	0.0019751	0.0004506582
	Buhu	0.000107704	0.0000489	0.0001800	0.0000427360

Source: Primary Data 2014

Table 3 shows the value of the intake of heavy metals in people who consume fish that contain heavy metals Hg. The highest intake values for Hg in the Huntulabohu village with an average daily intake are 0.000368241 mg/kg/day while the highest minimum value is equal to 0.0000788 villages Podutuma mg/kg/day and the maximum value for the highest Huntulabohu village also the daily intake value of 0.0019751 mg/kg/day.

Risk level (RQ)

 Table 4: Level of Risk (RQ) Oreochromis niloticus Consumption Contain Heavy Metals Hg with Exposure duration (Dt) 30

 Year of the community around Limboto Lake

Metal Type	Location	Mean	Min	Max	SD	
Risk level (RQ)						
Hg	Payunga	1.400156701	0.4356110	3.9827288	1.0504777054	
	Podutuma	1.005600534	0.7884445	1.5442192	0.2895360689	
	Ilomangga	0.117107499	0.0737973	0.2443288	0.0544173062	
	Kayubulan	1.379805788	0.6293542	2.8835866	0.5505958537	
	Huntulabohu	3.682414055	0.5422243	19.7506849	4.5065836498	
	Buhu	1.077039216	0.4891270	1.7995068	0.4273605138	

Source: Primary Data 2014

Table 4 shows the risk levels of heavy metals in the society who consume fish that contain heavy metals Hg. The highest value of the average level of risk (RQ) is 3.682414055 mg/kg/day in the Huntulabohu village and the lowest was found in the Ilmomangga village is 0.117107499 mg/kg/day, while the value of drinking level of risk (RQ) Podutuma village with the highest value level of risk (RQ) 0.788445 mg/kg/day while the maximum value of the highest in the Huntulabohu village the value of the level of risk of 19.7506849 mg/kg/day.

DISCUSSION

Mercury Limboto lake waters derived from mining activities, the results are based on interviews with people in the river, information from relevant agencies and direct field surveys known that the inlet and outlet are mining activities, in addition to the presence of heavy metals such as mekuri in Lake Limboto derived from natural. Mercury included in the pure elements in the form of granules in the midst of the rocks due to the effects of weather over a long time, first stone began to crack off a piece-by-piece later and eventually became the grains are refined. Together with rain water droplets will reach the water bodies and release positive ions.

The result of mercury concentration test showed that the concentration of mercury on the Nila Fish (Oreochromis niloticus) in Payunga Village was 0.0554 mg/kg, in Podutuma Village was 0.061 mg/kg, in Ilomangga Village was 0.007 mg/kg, in Kayubulan Village was 0.067 mg/kg, in Huntulabohu Village was 0.089 mg/kg, and in Buhu Village was 0.089 mg/kg. Besides, the result of the concentration of mercury on water in the lake test was about 0.0006 ppm until 0.0025 ppm. According to the SNI standard, the concentration of mercury on this Nila Fish still qualified because it was still \leq 0.5 mg/kg. Heavy metals mercury in Nila Fish (Oreochromis niloticus), can enter through the food they consume. The entry of mercury into the bodies of living organisms is primarily through food, because almost 90% of heavy metals or toxic substances enter the body through food materials (Palar, 2008). Nila fish are included in the class or omnivores eating everything, so it occupies the highest trophic waters. Mercury that enters the body and fish can be derived from water that

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has been contaminated with mercury, consumption sediment, aquatic plants, and small fish (Setianto, 2011).

Food lines, metallic mercury entered in two ways through the water (beverages) and plants (groceries). The amount of mercury that goes through a drink can be very high. The number could be many times compared to the amount of mercury that goes through the plant (Palar, 2008). Research on mercury levels and fish waters has been done about the analysis of mercury in water and Nila fish (Oreochromis niloticus) fish in the river Kaligarang Semarang. The results showed that the concentration of mercury in river water reached 0,005 ppm, and accumulates in fish tissue so that the concentration of mercury in fish reaching 0,014 ppm (Judge & Prayitno, 2003).

Amount of mercury entering through the water more into the bodies of living organisms, due to the heavy metals mercury in water may have experienced a doubling of the number of initial entry. The multiplicity of mercury in the water originated from the bacterial process of heavy metal ions contained in or deposition on mud bottom waters. This bacterial process can occur in all bodies of water (rivers and lakes) have conceded that mercury compounds, (Palar, 2008) so that it can be seen that the waters of Limboto Lake which is a habitat for fish, had been contaminated with mercury and lead contaminated fish also. Mercury that are waters of the lake, is absorbed by Nila fish (Oreochromis niloticus), thus accumulates in fish tissue and cause fish too polluted.

Based on the results of the analysis of mercury levels in the water, it is known that the highest mercury levels were in the village Kayubulan station 4, which is 0.0025 ppm while the lowest mercury levels were in the station 5 and 6 in the Village and the Village buhu Huntulabohu is 0.0006 ppm. Station 4 Kayubulan villages, there is a lake dredging activity, which may cause the element mercury naturally in rocks, soil and take apart and affect mercury levels around these waters. Additionally, Kayubulan village closes to the inlet of the lake, which is the river of Biyonga. Biyonga River is the main river carrying sediment, where the river Biyonga contributed 56% of the total sediment entering the lake (EART, 2009). In Biyonga upstream mining activity there, the activity that occurs in the body Biyonga River that is the excavation of sand, and the river flows through the area biyonga agriculture. The distance between the region's agricultural around in Biyonga River is up to Limboto Lake ± 10 kilometers.

Mercury compounding in agriculture is widely used as a fungicide. Because spraying is conducted openly and widely, so many other living organisms are exposed to these toxic compounds. That of the fungicide spraying not only kills the fungus but also other living organisms (Palar, 2008). The presence of mercury in fungsida strengthened with mercury toxicity case reports of outbreaks that occurred in Iraq in 1971-1972, in which more than 6,500 people were taken to the hospital due to methyl mercury poisoning and more than 450 people died, because eating bread from wheat has been preserved with a fungicide containing methyl-Hg (Darmono, 2010). Pesticides move from agricultural land into streams and lakes brought on by rainfall or evaporation left behind or late on the surface of the flow (Rahayu, et.al, 2009). But for the use of mercury-containing pesticides on agricultural biyonga river, needs to be more comprehensive study. In addition to passing through agricultural areas, river flow biyonga well before entering into Limboto Lake pass through residential areas, and hospitals.

Sediment can be caused due to erosion, the erosion in the river upstream of Limboto Lake inlet, caused by the destruction of forests. Where rainwater catchment watershed Bone Bolango Limboto has long been legalized turns into limited production forest that has pushed formally forest exploitation on a large scale. Forest area in the watershed Limboto is only 14 893 hectares (16:37% of the watershed area). Deforestation rates can increase the rate of soil erosion and cause the land is becoming critical. The rate of erosion in the watershed can Limboto reached 9,902,588.12 tons / year, or an average of 108.81 tons / ha / year and sedimentation in Limboto Lake of 0.438 mm / year (EART, 2009), so it is known that the incoming sediment in Limboto Lake also can contribute to mercury in the waters, where the element mercury contained naturally in rocks and soil in the sediment, carried in by rivers flow into Limboto Lake.

While low levels of mercury in the water, there are at stations 5 and 6 is on Buhu Villages and Huntulabohu Village. The presence of mercury in the station because the station is due to close to residential areas, so that the solid waste or leachate from the household can cause mercury pollution in the lake water. Where household waste, or leachate from waste, water can seep into the ground, so as to contribute mercury pollution. This is reinforced by studies on the relationship of pollution landfill leachate (landfill) waste Benowo

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with mercury levels in fish. The survey results revealed that there is so much mercury in the leachate at the outlet of waste water disposal plant (WWTP) Benowo landfill waste as much as 2.66 ppm) (Pragoyo & Sudarmaji, 2008).

The compounds used for the manufacture of mercury amalgam, paints, batteries, gold and silver extraction, dentures, anti-rust compound, photography and electronics. (Effendi, 2003) Thus, the household waste can also contribute to mercury pollution in rivers Alo-Pohu. The household waste is disposed around rivers and lakes can be either solid waste or liquid waste, solid waste, namely waste including paper, battery scrap, and waste plastics.

The level of risk (risk quotient [RQ]) is the amount of risk that is expressed in a number without a unit which is the intake calculation of the ratio between the dose / reference concentration of a non-carcinogenic risk agents and can also be interpreted as being safe / not safe an agent risks to organisms, system, or sub / populations. To determine the level of risk it is necessary to include the value of *the intake* to see how much of the daily intake of Oreochromis niloticus consumption containing heavy metals compared with the reference dose (Hg = 0.0001 mg / kg / day). The default value used exposure duration is 30 years.

RQ risk levels in Oreochromis niloticus are safe or not safe risk respondents from mercury exposure through fish consumption. The level of risk (RQ) is more than 1 mean that the fish is not safe for consumption by people around because the risk to cause health problems, so that risk management needs to reduce or minimize the risk of these health problems.

Based on the risk analysis carried out against respondents obtained that Payunga villagers who consume fish with high levels of Hg content of 0.0000554 mg / g for 30 years and the amount of intake 0.000132279 mg / kg / day had a risk level (RQ) 1:32, Podutuma villagers who consume fish with levels of Hg content of 0.000061 mg / g for 30 years and the amount of intake 0.000101436 mg / kg / day had a risk level (RQ) 1:01, to the village llomangga who consume fish with high levels of Hg content of 0.000007 mg / gram for 30 years and the amount of intake 1.1339 x 10⁻⁵ mg / kg / day had a risk level (RQ) 0:11, the people in the village Kayubulan who consume fish with high levels of Hg content of 0.000067 mg / gram for 30 years and the amount of intake 0.00013646 mg / kg / day had a risk level (RQ) 1:36, while in the Huntulabohu village who consume fish with high levels of 0.000089 mg / gram for 30 years and the amount of intake 0.00028658 mg / kg / day had a risk level (RQ) is 2.87, whereas in the Buhu village who consume fish with high levels of Hg content amount of intake 0.00010824 mg / kg / day had a risk level (RQ) 1:08.

Based on these values, it can be seen that the rate of population risk for mercury have RQ values> 1 mean a risk to the health of each respondent. Special to the llomongga village still is at safe limits, because the RQ value is still smaller than 1 (RQ <1). The risks referred to in this study is more probability means that the value of RQ> 1 would certainly not have health problems but the value is more indicative of the level of risk that someone greater than 1 will have a greater probabilities for the occurrence of a health effect compared with the value RQ \leq 1 The level of risk is influenced by the amount of intake of mercury that enters the human body. Intake in the body is affected by several factors, namely levels (concentration) of the chemical in the fish body weight, duration of exposure, the rate of intake, and frequency of exposure.

In the case of this study in which individuals with different weight but has a daily intake rate (R), frequency of exposure (FE) and exposure duration (Dt) are the same, have different risk. Where, the smaller the weight (Wb) someone then the level of risk (RQ) will also be greater. In addition, the level of risk (RQ) due to heavy metals in Nila fish (Oreochromis niloticus) fish will be growing greater with increasing duration of exposure (Dt) experienced by the individual. In this study, risk management in an effort to reduce the risk done in several ways: lower levels (Hg) Mercury, the Nila fish (Oreochromis niloticus) fish intake reduces the rate (R), reduce the frequency of exposure (FE) and reduce the exposure duration (Dt).

CONCLUSION

The result of this study showed that the concentration of mercury on Nila Fish in Limboto Lake was 0.007-0.089 mg/kg. Moreover, the average of the highest concentration of mercury on Nila Fish in Huntulabohu village and Buhu village was 0.089 mg/kg, meanwhile, the average of the concentration of mercury on water in the Limboto Lake was 0.0014 ppm. It means that Nila Fish, which was consumed by the

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people and water in the Limboto Lake have not exceeded the standard quality of SNI 2009 and Indonesian Government Regulation no. 82 of 2001. Besides, the average of risk level of fish consumption that contained mercury was more than 1 (RQ >1). Hence, it can be concluded that the consumption of Nila Fish (Oreochromis niloticus) which was taken from Limboto Lake was regarded unsafe to be consumed because it can cause health problems for the people. According to this study, the most effective risk management was by decreasing mercury (Hg) concentration and by controlling the rate of people's intake in consuming tilapia, which was taken from Limboto.

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